PHD

CHARACTERIZATION OF HYDRODYNAMICS PHENOMENA DURING DROP IMPACT ONTO DIFFERENT TYPES OF SUBSTRATES

The work was done under the supervision of Professor Arthur Soucemarianadin in the “Laboratoire des Ecoulements Géopgysiques et Industriels”, University of Grenoble (France).

The goal was to understand the mechanisms occurring during the impact of one or more drops of Newtonian fluid onto a flat solid and non porous substrate. Different parameters were studied including droplet velocity and size (from 30µm to 2mm), fluid surface tension and viscosity and wettability of the substrate. Experiments were performed on silicon surfaces where chemical patterning gave wide range of equilibrium contact angle. With distilled water, the range was from 5° to higher than 160°. An experimental set-up was specially constructed and it combined high speed cinematography and very short exposure times.

This work is relevant to different industrial applications including Ink Jet Printing and novel applications like *in situ* DNA synthesis.

Summary

Understanding drop impact phenomenon is important in industrial processes such as Ink Jet Printing but is a complex problem due to of the number of parameters. Droplet velocity and size, fluid viscosity and surface tension, wettability and roughness of the surface, etc....all have an effect on the impact of droplet onto a surface. Moreover, complexity also comes from the presence of a free surface, which shows high deformations, and the short time scale of the mechanism (insert see in figure 3).

Fluids used were a mixture of water and glycerol to vary the viscosity over two orders of magnitude with only a small change in surface tension. Model surfaces used all had exactly the same physical properties and a very low roughness (below 50Å). They were produced using silicon plates on to which chemical treatments were performed to reach equilibrium contact angles covering a wide range from hydrophilic (down to 5°) to super hydrophobic (greater than 160°) with distilled water. This technique was also used to create heterogeneous substrates with areas of different wettability.

Experiments were based on acquiring transient profiles during the impact process of one or several drops. In order to capture these transient deformations, an experimental set-up was built. This was based on a high speed camera (up to 90000fps with binning option, exposure time down to 2µs) and combined with light intensifier or laser diode. It provided exposure times down to tens of nanoseconds for micrometer drops and hundreds of nanoseconds for millimetre drop. Observations allowed the accurate capture of the transient high deformation of the droplet during the wetting process (figure 1). The excellent image quality obtained allowed droplet height, contact diameter and dynamic contact angle to be measured automatically using image processing algorithms specifically developed for the wetting mechanism.

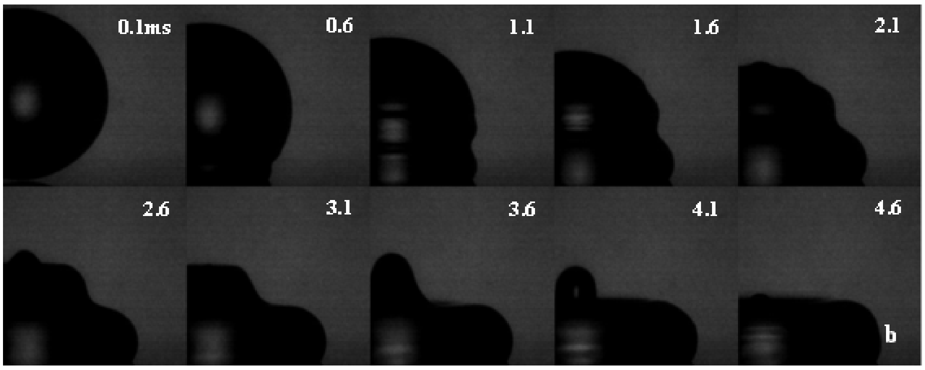


Figure 1: Transient deformations of the free surface during the first step of drop impact mechanism. Fluid: distilled water, $\theta_{eq} = 90°$.

The contact diameter was found to follow four different phases termed sequentially as kinematic, wetting, transition and capillary phase (figure 2). Three different behaviours were observed depending on the value of the maximum diameter (diameter reached at the end of wetting phase) compared with the final diameter (it only depends on the wettability of the surface). During to final phase of spreading, the contact diameter can show oscillations (low wettable surface) or can only spread under the action of surface tension. The prediction of the maximum diameter and its comparison with the final contact diameter is important in the aforementioned industrial applications.

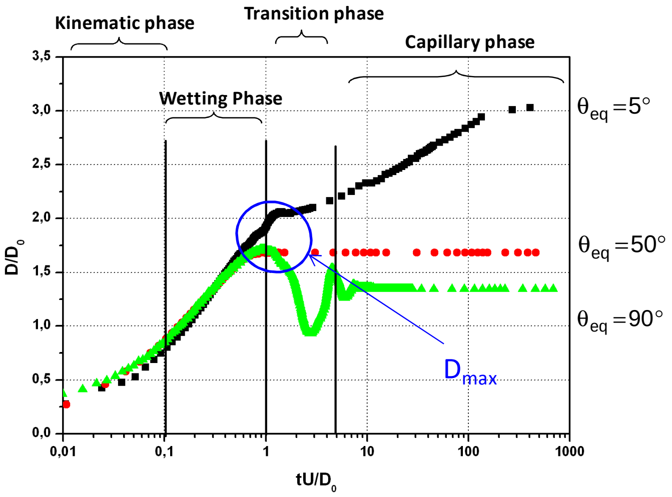
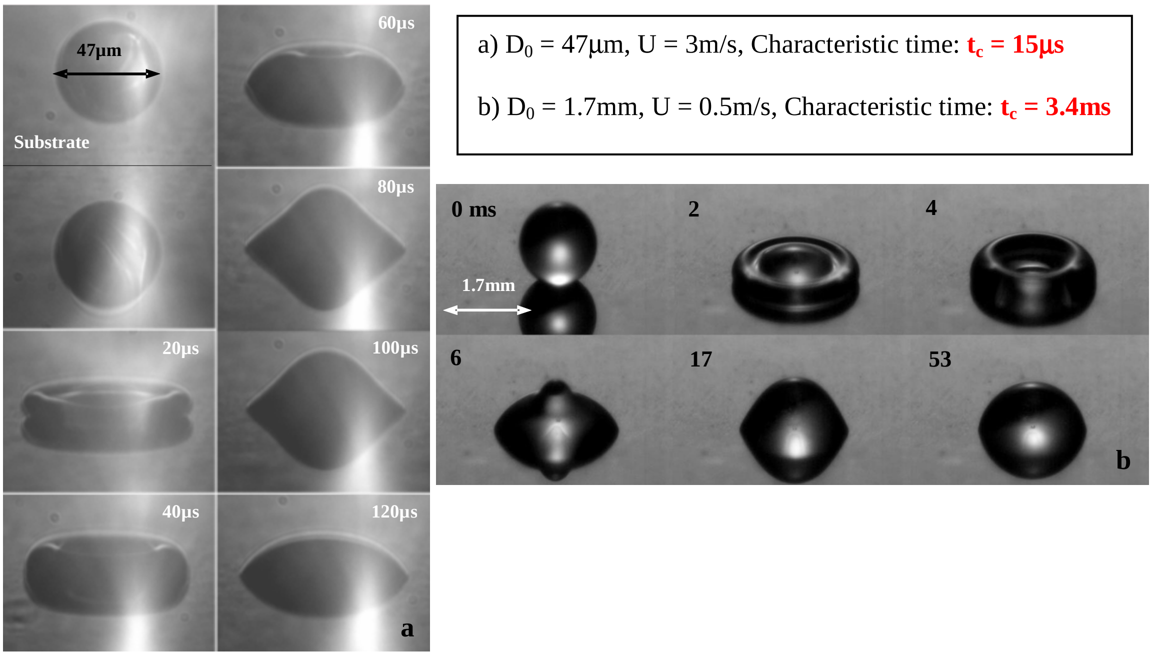


Figure 2: The three different behaviours of contact diameter during drop impact for three surfaces wettability. The four phases are reported on the curve, as the maximum diameter. Time and contact diameter are respectively dimensionless by initial diameter D_0 and characteristic time D_0/U.

The mechanism of drop impact is described by Navier-Stokes equations, and in their dimensionless form they show that the Weber (ration of inertia and surface tension, $\frac{\rho D_0 U^2}{\sigma}$) and Reynolds (ration of inertia and viscosity, $\frac{\rho D_0 U}{\mu}$) numbers control the wetting process. The characteristic time scale is the initial drop diameter D_0 (is also the characteristic length scale) divided by the impact velocity U. In theory, by maintaining We & Re numbers, the behaviour of the droplet is independent of size. An experimental comparison between millimetre and micrometer sized drops at comparable We & Re numbers is show below (figure 3a and 3b). Transient profiles appear to be experimentally similar.



a) D_0 = 47µm, U = 3m/s, Characteristic time: **t_c = 15µs**

b) D_0 = 1.7mm, U = 0.5m/s, Characteristic time: **t_c = 3.4ms**

Figure 3 : (a) Transient profiles of 47µm drop of industrial ink (σ = 33.2mN/m, μ = 3.2 mPa.s, ρ = 996 kg/m^3, U = 3m/s, D_0 = 47µm, We = 12.7, Re = 44). (b) Transient profiles of 1.7mm drop (mixture of distilled water, glycerol and surfactant, σ = 30mN/m, μ = 15 mPa.s, ρ = 1100 kg/m^3, U = 0.5m/s, D_0 = 1.7mm, We = 15.6, Re = 62).

For *in situ* DNA synthesis, substrates are chemically heterogeneous. Different cases were considered and it was demonstrated that, during kinematic and wetting phase, the droplet is only influenced by the surface on which it is spreading. Each part of the drop evolves on its own part of the substrate independently of the neighbouring regions development. Moreover, even if the substrate is divided in two different areas with different wettabilities, the spreading mechanism (in term of contact diameter and dynamic contact angle) on each part is similar to the spreading onto homogenous substrates with the same wettability.
Subsequently, a global motion of the drop, from the low wettable part of the substrate to the high wettable part of the substrate, by stick and slip, was observed due to the wettability gradient (figure 4). At the equilibrium state, the drop losses its axi-symetrical shape.

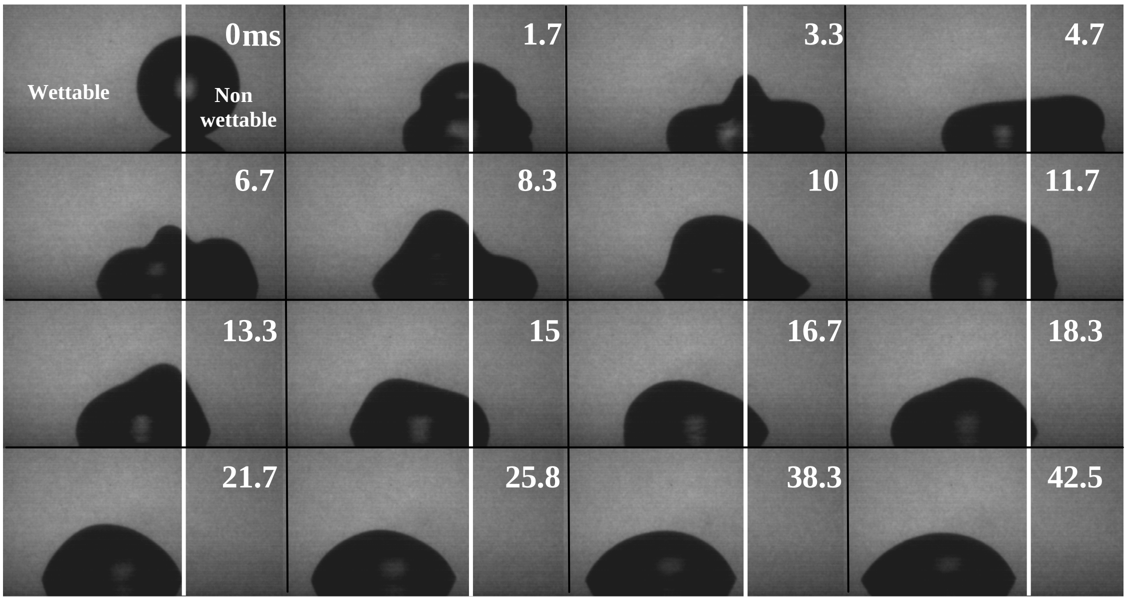


Figure 4: Distilled water drop impacting on wettable/non-wettable half plate, $\theta_w = 40°$, $\theta_{nw} = 90°$, $D_0 =$ 2.28mm, U = 0.3m/s, We = 2.8, Re = 684. White lines represent the separation of each part of the substrate.